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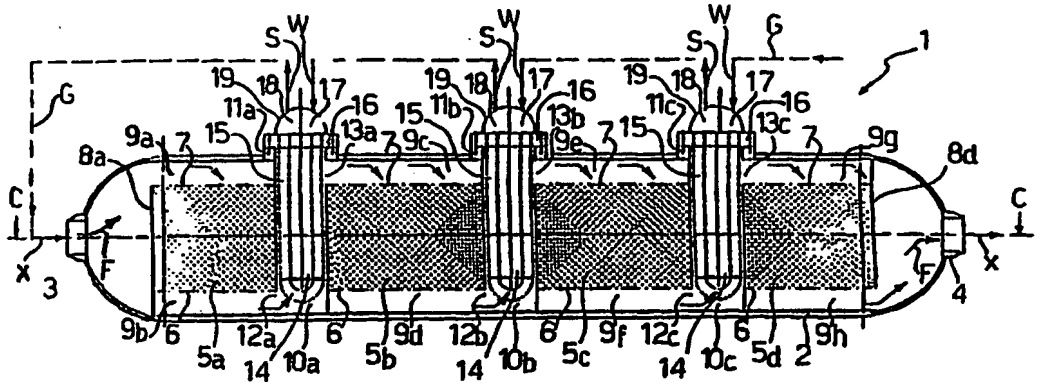

(57) Abstract

A horizontal reactor for heterogeneous exothermic synthesis, in particular for methanol synthesis, of simple construction and at the same time easy maintenance, comprises a substantially cylindrical external shell (2), a plurality of beds (5a-5d) arranged horizontally and side by side in said shell and comprising a lower gas-permeable wall (6) for gas outlet and at least one chamber (10a-10c) extending between at least two beds (5a-5d). Advantageously said chamber (10a-10c) is accessible from the outside and receives in a removable manner means (14) for cooling the gases flowing between the two side by side beds (5a-5d).

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Title: "Horizontal reactor for heterogeneous exothermic synthesis, in particular for methanol synthesis"

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DESCRIPTION

The present invention relates to a horizontal reactor for heterogeneous exothermic synthesis and in particular but not exclusively for methanol synthesis.

As known, in the field of heterogeneous exothermic synthesis in general, and more in particular in the production of methanol, the requirement for synthesis reactors having high productivity and conversion yield and at the same time low energy consumption and investment cost is increasingly felt.

Known art

For this purpose there has for some time been proposed as an alternative for the conventional vertical synthesis reactors with one or more catalytic beds the adoption of reactors with greater capacity arranged horizontally.

For example, in US patent US-A 4 696 799 there is described a horizontal reactor for heterogeneous exothermic synthesis of ammonia and comprising a cartridge in which are supported a plurality of catalytic beds in mutually spaced relationship.

In the cartridge are also provided cooling means comprising in particular two gas/gas heat exchangers for indirect cooling of the partially reacted synthesis gases flowing from the first and second catalytic beds.

More advantageous in many ways, the horizontal reactor

described above exhibits a series of drawbacks, the first of which is poor accessibility to all the internal parts of the reactor and in particular to the cooling means provided in the cartridge.

To perform any kind of maintenance operation on said means, e.g. replacement of defective or failed parts, it is necessary to remove the entire cartridge from the reactor, proceed with the required maintenance operations and lastly put the repaired cartridge back in the reactor.

As a consequence any work done on the reactor once it is in operation is long, difficult and especially costly.

Furthermore a reactor such as that just described comprises great structural complexity making practical construction difficult.

Because of these disadvantages, the horizontal reactors of the type considered have found rather limited use heretofore in the field of heterogeneous exothermic synthesis.

Summary of the invention

The technical problem underlying the present invention is to make available a horizontal reactor for heterogeneous exothermic synthesis having high productivity, high conversion yield, and low energy consumption and investment cost, which would be simple to construct and allow easy maintenance.

The technical problem is solved by a horizontal reactor for heterogeneous exothermic synthesis, in particular for methanol synthesis, comprising:

- an external shell of substantially cylindrical shape,

- a plurality of catalytic beds arranged horizontally and side by side in said shell and comprising a lower gas-permeable wall for gas outlet,

- at least one chamber extending between at least two of said beds and accessible from the outside of the shell,

- a fluid path for the gases flowing between said beds defined in said at least one chamber between opposed passages for gas inlet and outlet, and

- cooling means housed in a removable manner in said at least one chamber for indirect cooling of the gases flowing between said beds.

Advantageously, thanks to the special structure of the horizontal reactor in accordance with the present invention, it is now possible to accede easily to the cooling means, which can be easily replaced independently while avoiding all risk of damage to the other reactor parts.

In this manner the maintenance and repair operations on the cooling means are technically simple and economical.

Moreover, the arrangement of the removable cooling means in special chambers arranged between the catalytic beds and accessible from outside the shell also permits to carry out simply and effectively the maintenance of the catalytic bed, with particular reference to the loading and unloading of the catalyst.

Indeed, thanks to the presence of the above said chambers it is now possible to reduce the number of manholes and handholes necessary for such loading and unloading operations as compared with the known horizontal reactors.

Another advantage of the horizontal reactor according to

the present invention lies in its flexibility of use both as concerns the possibility of using interchangeably in the same reactor cooling means of different kinds, e.g. to produce high thermal level steam or for preheating the gaseous reagents supplied in the reactor, but also as concerns the possibility to vary the internal configuration of the reactor by housing the cooling means in the chambers in the desired sequence to perform or not indirect cooling between one bed and the other depending on the synthesis reaction it is intended to carry out.

In the latter case the shell is advantageously provided with removable covers arranged in correspondence of the chambers not occupied by cooling means.

Advantageously at least two and preferably all the catalytic beds arranged side by side in the shell are mutually adjacent.

In this manner there is achieved optimal utilization of the useful volume of the reactor with resulting high filling degree.

It is also found that for optimal heat exchange between the partially reacted hot gases and the cooling fluid the chamber in which are housed the cooling means is preferably cylindrical in shape.

In a special and advantageous embodiment of the horizontal reactor of the present invention the cooling means comprise a boiler for high pressure steam generation or a gas/gas heat exchanger for preheating the gaseous reagents supplied to the reactor.

In the first case there is achieved advantageously a recovery at high thermal level of the reaction heat to generate the high pressure steam usable in other parts of

the plant.

The characteristics and advantages of the heterogeneous exothermic synthesis reactor according to the present invention are set forth in the description of an embodiment thereof given below by way of non-limiting example with reference to the annexed drawings.

Brief description of the drawings

In the drawings:

FIG. 1 shows a plan view of a horizontal reactor for heterogeneous exothermic synthesis in accordance with the present invention,

FIG. 2 shows a longitudinal cross section view of the horizontal reactor of FIG. 1, taken along lines A-A of FIG. 1,

FIG. 3 shows a cross section view of the horizontal reactor of FIG. 1, taken along lines B-B of FIG. 1, and

FIG. 4 shows another longitudinal cross section of the horizontal reactor of FIG. 1, taken along lines C-C of FIG. 2.

Detailed description of a preferred embodiment

With reference to figures 1-4 reference number 1 indicates as a whole a horizontal reactor for heterogeneous exothermic synthesis and in particular for methanol synthesis.

The reactor 1 comprises and external tubular shell 2 fitted at its ends with openings 3, 4 respectively for inlet of reagent gases and outlet of the reaction products.

5a-5d indicate a plurality of catalytic beds arranged

horizontally and side by side in the shell 2, along a longitudinal axis x-x of the reactor 1, and comprising opposed gas permeable walls 6, 7 respectively lower and upper.

The upper gas permeable wall 7 has the function of avoiding an excessively violent impact of the gaseous reagents on the catalyst. Said wall is therefore not required in the case of a low-velocity gas flow. The gas-permeable wall 6 has the function of supporting the catalytic mass.

The beds 5a-5d are adjacent and mutually separated by a plurality of baffles, all indicated by 8, extending transversally to the longitudinal axis x-x of the reactor 1. 8a and 8d indicate opposed closing side walls of the catalytic beds 5a and 5d respectively.

A plurality of air spaces 9a-9h are defined between the inner wall of the shell 2 and the gas-permeable walls 6 and 7 of the beds 5a-5d for passage of the gases from one catalytic bed to the other.

Between the catalytic beds 5a-5d is defined a plurality of cylindrical chambers 10a, 10b, 10c extending transversally to the beds and accessible from the outside of the shell 2 through respective apertures 11a, 11b, 11c.

Each of said chambers 10a, 10b, 10c is equipped with opposed passages 12a, 13a to 12c, 13c for inlet and outlet of gas respectively, which open in correspondence of the air spaces 9a-9h. A plurality of fluid paths is defined in this manner between pairs of adjacent beds 5a-5d.

In FIG. 2 inside the chambers 10 are housed in a removable manner respective cooling means 14 for the indirect cooling of the gases flowing between said adjacent beds 5a-5d.

Said cooling means 14 comprise a plurality of U-shaped tubes 15 extending into the chambers 10a-10c. The ends of the tubes 15 are in fluid communication through a tube plate 16 with respective chambers 17, 18 for inlet and outlet of cooling fluid.

The chambers 17, 18 are made in a cover 19 of the cooling means 14 which is integral with the tube plate 16.

The tube plate 16 is fixed in a conventional removable manner to the shell 2 in correspondence of the apertures 11a-11c, e.g. by bolting.

In a preferred embodiment said cooling means 14 use water as cooling fluid and therefore constitute essentially a boiler for high thermal level steam generation

As an alternative the means 14 can use as cooling fluid a part of the cold reagent gases, thus constituting a preheater for the reagent gases.

In another embodiment (not shown) the boiler or heat exchanger can be of the bayonet type.

The flow of cooling fluid through said cooling means 14 can be of the natural circulation type or of the forced circulation type.

In FIGS. 2 and 3 the arrows F indicate the various paths taken by the gaseous reagents along the air spaces 9a-9h through the catalytic beds 5a-5d and the cooling means 14.

In FIG. 2 the arrows W and S indicate respective paths for the cooling water entering the chambers 17 and the steam leaving the chambers 18.

The broken-line arrows G indicate the path of the cold synthesis gas preheated by the means 14 in the preheater

version.

Operation of the horizontal reactor of the present invention is as follows.

With reference to FIG. 2 a flow of reagent gases, e.g. H_2, CO and CO_2 already preheated to reaction temperature (approximately 240 °C) is let into the reactor 1 through the opening 3 for gas inlet, flows along the air space 9a and traverses adiabatically the first catalytic bed 5a from the top downward.

The partially reacted gases come out of the latter at a temperature of approximately 290 °C and enter through the passage 12a into the chamber 10a where they are cooled by indirect thermal exchange with a flow of water passing through the tubes 15 of the boiler 14.

In this manner the synthesis gas is brought to the initial temperature with simultaneous high pressure steam generation, e.g. 20-25 bar. The steam thus produced can be advantageously used, e.g. in the methanol distillation phase.

The cooled flow of partially reacted gas leaving the chamber 10a is collected in the air space 9c an then enters the second bed 5b in a manner analogous to that described above.

The reaction products coming from the last bed 5d come out of the reactor through the opening 4.

The horizontal reactor thus conceived has optimal thermodynamic efficiency comparable in the methanol synthesis field to that of an isotherm reactor of equal catalytic volume which is known to allow achievement of the highest conversion yield.

Advantageously the horizontal bed according to the present invention having a very simple internal structure and of modular type can contain a variable number of catalytic beds of at least a minimum of two intercalated with cooling means, compatibly with the internal pressure conditions in the shell.

The preferred embodiments are those with four adiabatic catalytic beds with three intermediate cooling means (as in the example shown), or three adiabatic catalytic beds and two intermediate cooling means.

In addition, thanks to the presence of catalytic beds arranged side by side and at least partly adjacent, it is possible to achieve a high degree of utilization of the useful volume of the reactor, which can be occupied even up to 80-83% by the catalyst.

Consequently, for equal installed catalyst volume the reactor according to the present invention will have a smaller pressure body (shell) than that of a reactor of the prior art, with considerable material economy.

It will be appreciated that the horizontal reactor in accordance with the present invention is advantageously applicable for heterogeneous exothermic synthesis of different types compatibly with the reactor structure.

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The above description shows clearly the numerous advantages achieved by the horizontal reactor for heterogeneous exothermic synthesis in accordance with the present invention, including:

- a structurally simple reactor easy to construct, practical and flexible in use,

- extremely easy maintenance,

- optimal utilization of the useful volume of the reactor,

- recoverability of reaction heat at high thermal level producing high pressure steam, and

- high thermodynamic yield.

CLAIMS

1. Horizontal reactor for heterogeneous exothermic synthesis, in particular for methanol synthesis, comprising:

- an external shell (2) of substantially cylindrical shape,

- a plurality of catalytic beds (5a-5d) arranged horizontally and side by side in said shell (2) and comprising a lower gas-permeable wall (6) for gas outlet,

- at least one chamber (10a-10c) extending between at least two of said beds (5a-5d) and accessible from the outside of the shell (2),

- a fluid path for the gases flowing between said beds (5a-5d) defined in said at least one chamber (10a-10c) between opposed passages (12a,13a-12c,13c) for gas inlet and outlet, and

- cooling means (14) housed in a removable manner in said at least one chamber (10a-10c) for indirect cooling of the gases flowing between said beds (5a-5d).

2. Reactor according to claim 1 characterized in that at least two of said beds (5a-5d) are adjacent.

3. Reactor according to claim 1 characterized in that it also comprises a plurality of air spaces (9a-9h) for passage of the gases from and to said beds (5a-5d) defined between the internal wall of the shell (2) and opposed upper and lower surfaces (7,6) of said beds.

4. Reactor according to claim 3 characterized in that said passages (12a,13a-12c,13c) for gas inlet to and outlet from said at least one chamber (10a-10c) are open in correspondence of said air spaces (9a-9h).

5. Reactor according to claim 1 characterized in that said at least one chamber (10a-10c) is substantially cylindrical in shape.

6. Reactor according to claim 1 characterized in that said cooling means comprise at least one boiler (14) for high thermal level steam generation.

7. Reactor according to claim 6 characterized in that said boiler (14) is of the tube nest or bayonet type.

8. Reactor according to claim 7 characterized in that said boiler (14) is of the natural or forced circulation type.

9. Reactor according to claim 1 characterized in that said cooling means comprise at least one gas/gas heat exchanger (14).

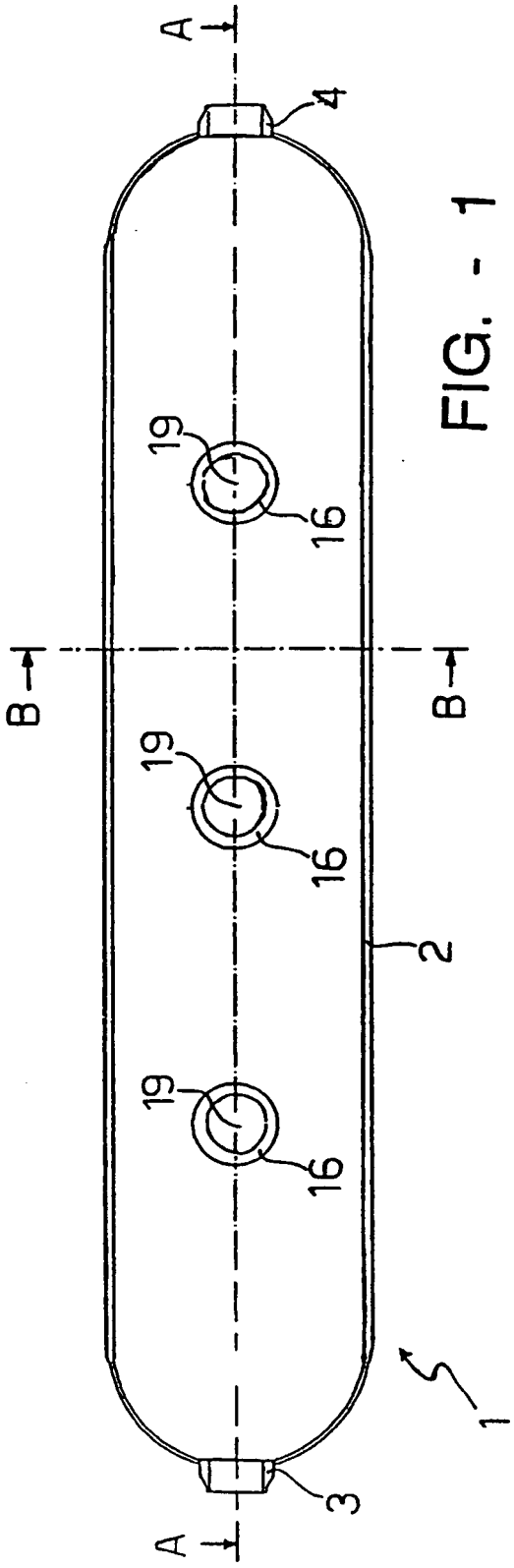


FIG. - 1

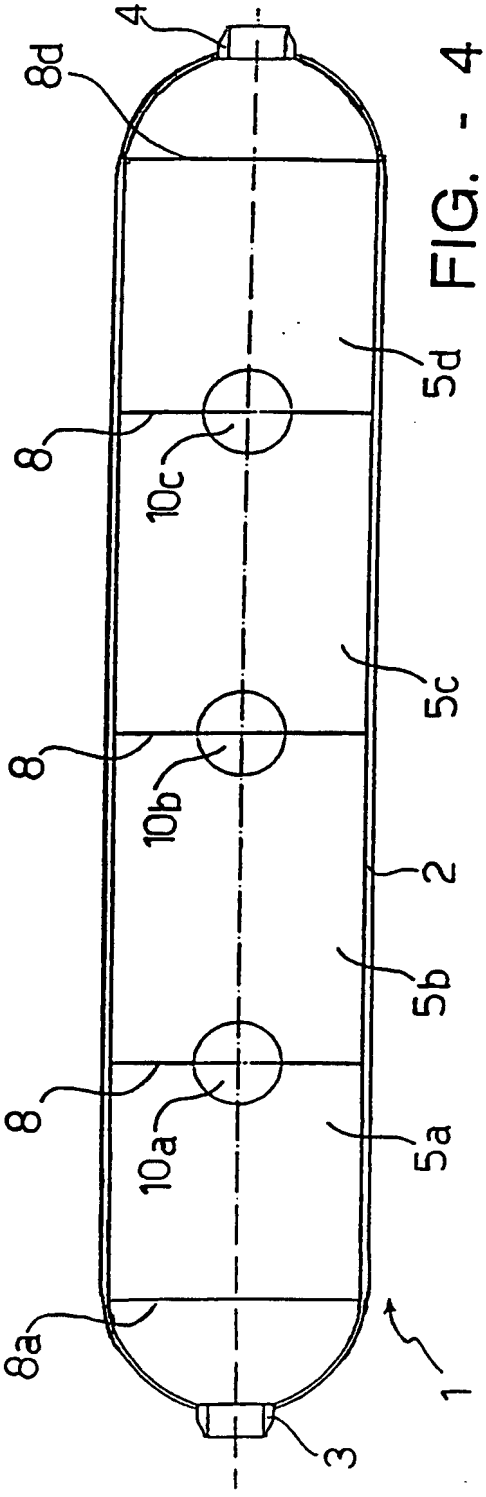
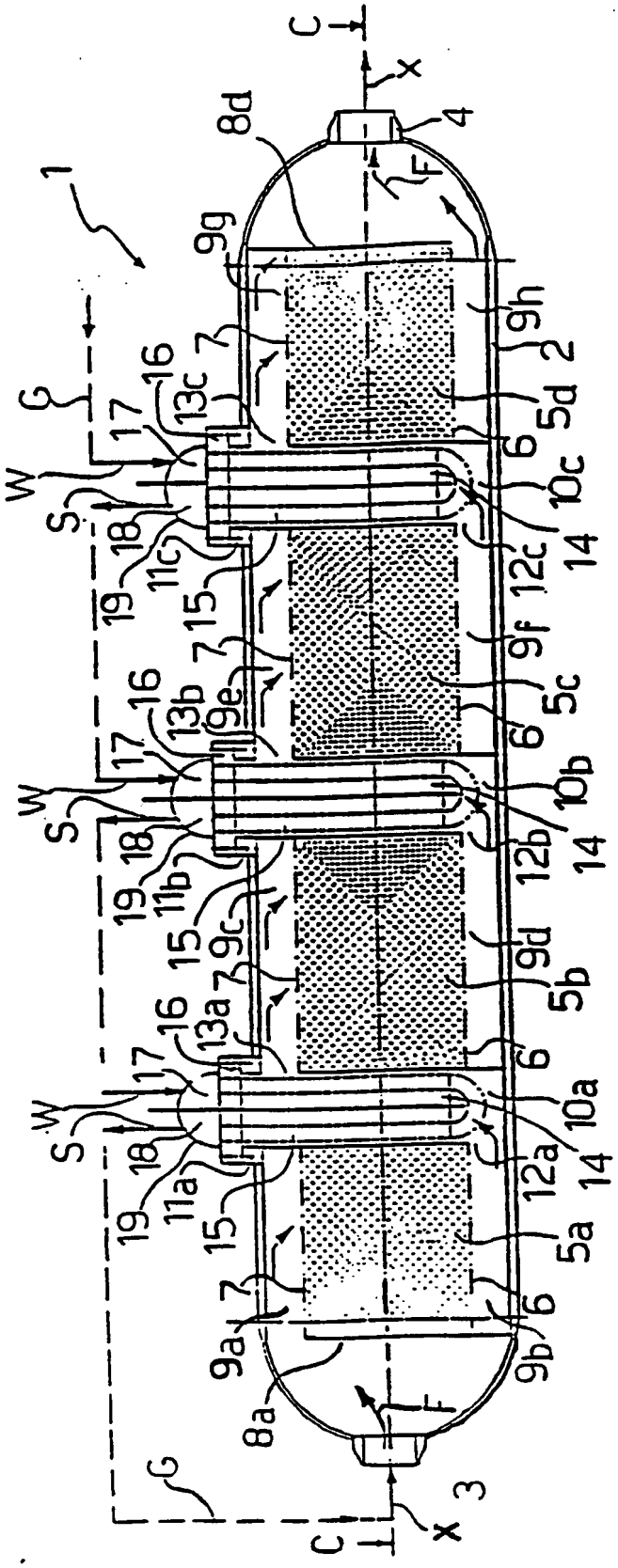


FIG. - 4


1
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3
4
5a
5b
5c
5d
6
7
8a
8d
9a
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9c
9d
9e
9f
9g
9h
10a
10b
10c
11a
11b
11c
12a
12b
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19
C
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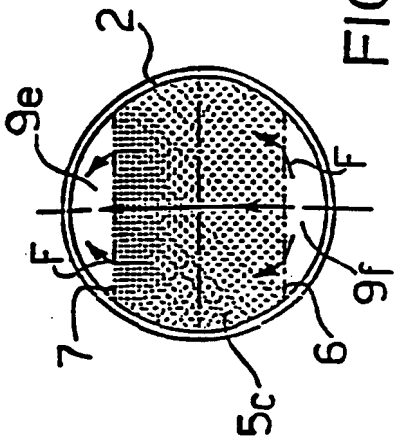
S
W
X

FIG. - 2


2
5c
6
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9e
9f
F
X

FIG. - 3

INTERNATIONAL SEARCH REPORT

:rnational Application No
PCT/EP 95/00938

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B01J8/04 B01J8/02 C07C31/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 B01J C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE,A,24 49 789 (DEGGENDORFER WERFT UND EISENBAU GMBH) 11 March 1976 see column 1, line 39 - line 47 see column 3, line 2 - line 37 see column 4, line 40 - column 5, line 3 see figures 1,8	1-4,7
A	GB,A,670 299 (METALLGESELLSCHAFT AKTIENGESELLSCHAFT) 16 April 1952 see page 1, line 10 - line 21 see page 1, line 71 - page 2, line 57 see page 4, line 57 - line 98 see page 5, line 72 - line 91 see figures 2-4	1,2,4-6

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	FR,A,2 138 701 (OSTERREICHISCHE STICKSTOFFWERKE AG) 5 January 1973 see page 1, paragraph 1 see page 2, line 4 - page 4, line 14 see figure	1-5,9
A	US,A,4 696 799 (THE M.W.KELLOGG COMPANY) 29 September 1987 cited in the application see the whole document	1-5,9
A	FR,A,2 182 009 (INSTITUTUL DE PROIECTARI PENTRU INDUSTRIA CHIMICA ANORGANICA) 7 December 1973 see page 5, line 20 - line 35	6-8
A	DE,A,33 10 772 (INTERATOM INTERNATIONALE ATOMREAKTORBAU GMBH) 27 September 1984 see the whole document	1,2,5,6

INTERNATIONAL SEARCH REPORT

Information on patent family members

ernational Application No
PCT/EP 95/00938

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
DE-A-2449789	11-03-76	FR-A-	2330451	03-06-77
		JP-A-	51064476	03-06-76
		US-A-	4099926	11-07-78
GB-A-670299		NONE		
FR-A-2138701	05-01-73	AT-B-	306745	15-03-73
		BE-A-	783830	23-11-72
		DE-A-	2126211	21-09-72
		GB-A-	1387044	12-03-75
		NL-A-	7205262	28-11-72
US-A-4696799	29-09-87	AU-B-	582088	09-03-89
		AU-A-	7557987	21-01-88
		CA-A-	1272868	21-08-90
		DE-A-	3772294	26-09-91
		EP-A,B	0256299	24-02-88
		NO-B-	176210	14-11-94
FR-A-2182009	07-12-73	DE-A-	2320614	15-11-73
		US-A-	3851406	03-12-74
DE-A-3310772	27-09-84	NONE		